

117 N. Division Ann Arbor, Michigan 48104 734.761.3186 734.663.2414 (fax)

September 28, 2005

Dr. Michael D. Shelby CERHR Director NIEHS, PO Box 12233, MD EC-32 Research Triangle Park, NC 27709

Dear Dr. Shelby and NTP Expert Panel,

We would like to provide additional information to the panel regarding the use and exposure to di (2-ethylhexyl) phthalate (DEHP)'s. The draft expert panel report did not adequately assess all environments in which people are exposed to phthalates. We particular find that the study fails to assess exposure from the transportation sector, in particular automobiles and light trucks. Interior auto cabin phthalate and DEHP exposure both from air and dust is significant and should be included in the expert panel report.

We have attached a brief summary of the literature on DEHP in automobiles, as well as, summaries of data from recent sampling of auto interiors completed by the Ecology Center. Please fee free to contact us if you have any questions.

Sincerely,

Jeff Gearhart, M.S. Ecology Center Hans Posselt, Ph.D. Ecology Center

Overview of Phthalic Acid Esters in Air, Dust and Window Films of Passenger Vehicles

Phthalic acid esters (phthalates) are among the most ubiquitous synthetic chemicals in the environment¹ and are nearly always found at some concentration in virtually all people and wildlife.² Phthalates are found in the air and dust in homes and offices,³ and on indoor and exterior window films of homes and office buildings^{4,5}.

Phthalates are predominantly used as plasticisers in soft plastics, such as in a large variety of polyvinyl chloride (PVC) products including seat covers and interior trim in vehicles. Global production of phthalates is an estimated 3.5 million metric tons per year,⁶ of which 80-90 percent is used as additives in flexible PVC. ⁷ Roughly 50 percent of the market share for phthalates is accounted for by diethylhexyl phthalate (DEHP),⁸ at least 95 percent of which is added to PVC to give it flexibility.⁹ As noted by Koch et. al., "This is of greatest importance for public health since DEHP is not only the most important phthalate with respect to its production, use and occurrence and omnipresence but also the phthalate with the greatest endocrine disrupting potency."¹⁰

While environmental releases of industrial chemicals are most commonly associated with their manufacture and disposal, it is estimated that more than 75 percent of phthalate releases to the environment occur during the use of products that contain phthalates. ¹¹ Clausen et al. (2004) documented releases to air of DEHP from PVC flooring. ¹² In studying phthalate emissions from PVC skirting, PVC flooring, and other materials, Afshari et al. concluded as follows: ¹³

"Plasticizers used in surface materials indoors can be detected in the indoor air and human exposure to plasticizers can be expected. This study shows that the concentration of phthalates in indoor air is independent of ventilation rates and the area of surface materials containing plasticizers, i.e. a small area of plasticizer containing products emits almost as much as a large area. Therefore, if the surface materials contain plasticizers, it is impossible to avoid the phthalates in indoor air."

Phthalates are among the most ubiquitous synthetic chemicals in the environment¹⁴ and are nearly always found at some concentration in virtually all people and wildlife.¹⁵ Phthalates are found in the air and dust in homes and offices.¹⁶ Exposure to phthalates and their metabolites have been associated with a broad range of health effects: ^{17,18, 19, 20, 21, 22,23}

- asthma and other respiratory problems, rhinitis and eczema in children;
- premature breast development in female children; and
- deteriorated semen quality, low sperm counts, and poor sperm morphology in men.

The daily intake of phthalates frequently exceeds the EPA reference dose and is of great environmental and health concern. 24

In Europe, DEHP is classified as "toxic to reproduction." Likewise, in California, DEHP is listed as potential carcinogen in the chemical list of Proposition 65.²⁵

Of concern to occupants of the vehicles is exposure to phthalates. A phthalate commonly used in PVC, di-2-ethylhexyl phthalate (DEHP) is a reproductive toxicant -- i.e., damages the reproductive system of animals. The heating of PVC by the sun increases the rate at which DEHP leaks out of the plastic.

Research shows that as the temperature of a vehicle's interior rises, the concentration of DEHP in the air rises. As detailed in Table 1, the levels of DEHP in automotive interiors can rise above the US Environmental Protection Agency's Reference Dose (RfD) of $20 \mu g/kg$ bw/day for

DEHP.¹ While no one sits in a vehicle at 140°F, we do enter into vehicles that have baked in the sun and reached temperatures much higher than 77°F.

Table 1: DEHP Exposures from Vehicle Interiors²⁶

Temperature in a Vehicle Interior	DEHP Levels Measured (nanogram per cubic meter ng/m³)	Human Exposure (microgram per kilogram body weight per day µg/kg bw/day)
77°F (or 25°C) 140°F (or 60°C)	10,000 ng/m ³ 300,000 ng/m ³	<1 µg/kg bw/day 30 µg/kg bw/day
Highest documented level of DEHP in vehicles	1,000,000 ng/m ³	100 μg/kg bw/day
US EPA Reference Dose (RfD)¹ for DEHP		20 µg/kg bw/day it risk of harmful non-cancer effects during a lifetime.

The CERHR draft cites DEHP air concentrations 109 ng/m3 (median 55, range 20-240)(Table 1, page 2). Potential air concentrations cited in Table 1 are significantly higher then those cited in the CERHR draft report and show autos to be previously not considered, significant source of DEHP exposure.

Tables 2 and Table 3 show the concentrations of phthalates found in recently completed automotive dust samples and window film samples respectively (Ecology Center, in press).

Table 2: Concentration of Phthalates (mg/g) in Vehicle Dust

DMP	DEP	DPP	DBP	DIBP	BBP	DEHP	DOP	Total
<dl< th=""><th><dl< th=""><th><dl< th=""><th>3</th><th>1</th><th>6</th><th>49</th><th>4</th><th>63</th></dl<></th></dl<></th></dl<>	<dl< th=""><th><dl< th=""><th>3</th><th>1</th><th>6</th><th>49</th><th>4</th><th>63</th></dl<></th></dl<>	<dl< th=""><th>3</th><th>1</th><th>6</th><th>49</th><th>4</th><th>63</th></dl<>	3	1	6	49	4	63

<dl = below detection limit

The CERHR draft cites DEHP concentrations in dust of 3.24 (20-240) g/kg. Auto dust concentrations cited in Table 2 are in this range as well and should be cited as another source of ongoing exposure.

Table 3: Concentration of Phthalates (mg/m2) in Vehicle Window Films

	DMP	DEP	DPP	DBP	DIBP	BBP	DEHP	DOP	l otal ug/m2	
Parked vehicles(1)	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td>10</td><td>21</td><td>8</td><td>39</td><td></td></dl<></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td><dl< td=""><td>10</td><td>21</td><td>8</td><td>39</td><td></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td><dl< td=""><td>10</td><td>21</td><td>8</td><td>39</td><td></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td>10</td><td>21</td><td>8</td><td>39</td><td></td></dl<></td></dl<>	<dl< td=""><td>10</td><td>21</td><td>8</td><td>39</td><td></td></dl<>	10	21	8	39	
In Use Vehicles(2)	<dl< td=""><td><dl< td=""><td><dl< td=""><td>3</td><td><dl< td=""><td>2</td><td>7 (3-24)</td><td>1</td><td>9</td><td></td></dl<></td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td>3</td><td><dl< td=""><td>2</td><td>7 (3-24)</td><td>1</td><td>9</td><td></td></dl<></td></dl<></td></dl<>	<dl< td=""><td>3</td><td><dl< td=""><td>2</td><td>7 (3-24)</td><td>1</td><td>9</td><td></td></dl<></td></dl<>	3	<dl< td=""><td>2</td><td>7 (3-24)</td><td>1</td><td>9</td><td></td></dl<>	2	7 (3-24)	1	9	
(1) 10 vehicles from a parking lot.										
(2) Vehicles which had been driven for an undetermined										
time. Sampled at a recycling drop-off center.										

The CERHR draft does not cite any data on phthalates or DEHP on window films. However, the Ecology Center recently collected data on DEHP concentration on front windshields of over 100 vehicles. The results are summarized in Table 3 above.

Diethylhexyl phthalate (DEHP) is by far the predominant representative of this group in both dust and film samples. The highest total phthalate concentration was found in the wipe-composite representing parked vehicles indicating a build-up of film deposits during parking. This corresponds well with data on air concentrations (Table 1) which indicate significantly higher levels of DEHP when vehicles are not in use and exposed to solar heating.

¹ The RfD is an estimate of the daily oral exposure to a chemical that is likely to be without risk of harmful non-cancer effects during a lifetime.

__

² Health Care Without Harm, 2002. Aggregate Exposures to Phthalates in Humans. Arlington, VA: Health Care Without Harm. www.noharm.org/details.cfm?type=document&id=796

³ Rudel, R.; Camann, D.; Spengler, J.; Korn, L.; Brody, J. 2003 Phthalates, alkylphenols, pesticides, polybrominated diphenyll ethers, and other endocrine-disrupting compounds in indoor air and dust. *Environ. Sci. Technol.* 37: 4543-4554.

⁴ Butt, C.M.; Diamond, M.L.; Truong, J.; Ikonomou, M.G.; ter Schure, A.F.H. 2004 Spatial Distribution of Polybrominated Diphenyl Ethers in Southern Ontario as Measured in Indoor and Outdoor Window Organic Films. *Environmental Science and Technology* 38: 724-731.

⁵ Rudel, R.A.; Camann, D.E.; Spengler, J.D.; Korn, L.R.; & Brody. J.G. 2003 Phthalates, alkylphenols, pesticides, polybrominated diphenyl ethers, and other endocrine d-disrupting compounds in indoor air and dust. *Environmental Science & Technology* 37(20): 4543.

⁶ Bornehag, C.-G.; Sundell, J.; Weschler, C.; Sigsgaard, T.; Lundgren, B.; Hasselgren, M.; Hagerhed-Engman, L. 2004 The association between asthma and allergic symptoms in children and phthalates in house dust: A nested case–control study. *Environ Health Perspect* 112: 1393 – 1397

⁷ Bizzari, S.; Gubler, R.; Kishi, A. 2003 CEH Report: Plasticizers.

http://ceh.sric.sri.com/Public/Reports/576.0000/. Accessed February 4, 2005.

⁸ Bornehag, C.-G.; Sundell, J.; Weschler, C.; Sigsgaard, T.; Lundgren, B.; Hasselgren, M.; Hagerhed-Engman, L. 2004 The association between asthma and allergic symptoms in children and phthalates in house dust: A nested case–control study. *Environ Health Perspect* 112: 1393 – 1397

⁹ Langenkamp, H.; Marmo, L. (eds) 2000. Proceedings: Workshop on Problems Around Sludge, 18-29 November 1999, Stresa, Italy. EUR 19657 EN, European Commission Joint Research Center.

¹⁰ Koch, H.; Drexler, H.; & Ångerer, J. 2003 An estimation of the daily intake of di(2-ethylhexyl)phthalate (DEHP) and other phthalates in the general population. *International Journal of Hygiene and Environmental Health* 206: 77-83.

¹¹ Kenny, A.; Thomas, K.; Peeler, E.; Osborn, D.; Waldock, M.; Matthiessen, P.; Shore, R.; Wright, J.; and Feist, S. 2002 Assessing the potential of environmental monitoring for detecting and responding to damage caused by the use of chemicals. Report by the Centre for Environment, Fisheries and Aquaculture Science (CEFAS) for the Royal Commission on Environmental Pollution.

¹² Clausen, P.; Hansen, V.; Gunnarsen, L.; Afshari, A.; Wolkoff, P. 2004 Emission of Di-2-ethyhexyl phthalate from PVC flooring into air and uptake into dust: emission and sorption experiments in FLEC and CLIMPAO. *Environ. Sci. Technol.* 38: 2531-2537

¹³ Afshari, A.; Gunnarsen, L.; Clausen, P.; Hansen, V. 2004. Emission of phthalates from PVC and other materials. *Indoor Air* 14: 120-128.

¹⁴ Dorey, C. 2003. Chemical Legacy Contamination of the Child. London: Greenpeace UK

¹⁵ Health Care Without Harm, 2002. Aggregate Exposures to Phthalates in Humans. Arlington, VA: Health Care Without Harm. www.noharm.org/details.cfm?type=document&id=796

¹⁶ Rudel, R.; Camann, D.; Spengler, J.; Korn, L.; Brody, J. 2003 Phthalates, alkylphenols, pesticides, polybrominated diphenyll ethers, and other endocrine-disrupting compounds in indoor air and dust. *Environ. Sci. Technol.* 37: 4543-4554.

¹⁷ Jaakkola, J.J.K.; Øie, L.; Nafstad, P.; Botten, G.; Lødrup-Carlson, K.C.; Samuelsen, S.O. and Magnus, P. 1999 Interior surface materials in the home and the development of bronchial obstruction in young children in Oslo, Norway, *Am. J. Public Health*, 89, 188–192.

¹⁸ Øie, L.; Hersoug, L.G. and Madsen, J.Ø. 1997 Residential exposure to plasticizers and its possible role in the pathogenesis of asthma, *Environ. Health Perspect.* 105, 972–978.

¹⁹ Bornehag, C.-G.; Sundell, J.; Weschler, C.; Sigsgaard, T.; Lundgren, B.; Hasselgren, M.; Hagerhed-Engman, L. 2004 The association between asthma and allergic symptoms in children and phthalates in house dust: A nested case-control study. *Environ. Health Perspect.* 112:1393–1397.

²⁰ Colon, I.; Caro, D.; Bourdony, C. J. & Rosario, O. 2000 Identification of phthalate esters in the serum of young Puerto Rican girls with premature breast development. *Environmental Health Perspectives* 108(9): 895-900

²¹ Duty, S.; Silva, M.; Barr, D.; Brock, J.; Ryan, L.; Zuying, C.; Herrick, R.; Christiani, D.; Hauser, R. 2002 Urinary phthalate monoesters at general populations exposure levels are associated with altered semen quality. *Epidemiology* July 2002, Volume 13 Number 4 Supplement ²² Duty, S.; Singh, N.; Silva, M.; Barr, D.; Brock, J.; Ryan, L.; Herrick, R.; Christiani, D.; Hauser, R. 2003

²⁴ Duty, S.; Singh, N.; Silva, M.; Barr, D.; Brock, J.; Ryan, L.; Herrick, R.; Christiani, D.; Hauser, R. 2003 The relationship between environmental exposures to phthalates and DNA damage in human sperm using the Neutral Comet Assay. *Environ. Health Perspect.* 111: 1164-1169.

¹ Dorey, C., 2003. Chemical Legacy Contamination of the Child. London: Greenpeace UK

²³ Rozati, R.; Reddy, R.; Reddanna, P.; Mujtaba, R. 2002 Role of environmental estrogens in the deterioration of male factor fertility. *Fertility & Sterility* 78: 1187-1194

²⁵ Office of Environmental Hazard Assessment (OEHHA); Proposition 65; List of Chemicals, Regulatory Update 2001: http://www.calprop65.com/01regs.html

²⁴ Koch, H.; Drexler, H.; & Angerer, J. 2003 An estimation of the daily intake of di(2-ethylhexyl)phthalate (DEHP) and other phthalates in the general population. *International Journal of Hygiene and Environmental Health* 206: 77-83.

²⁶²⁶ Huber, et.al., Huber, W.W., B. Grasl-kraupp, and R. Schulte-Hermann, 1996. "Hepatocarcinogenic potential of di(2-ethylhexyl)phthalate in rodents and its implications on human risk." *Critical Reviews in Toxicology*, 26(4):365-481.